Anharmonic Interactions Between Novel Surfaces and Adsorbates

Final Technical Report

Summary

This grant has supported the salary and research expenses of two post-doctoral fellows, Dirk Porezag and Jens Kortus, and a short-term visit by Alexander Sieck, then a graduate student at the Technical University of Chemnitz. Work was completed on three projects: the development of a scheme for optimizing Gaussian basis sets to be used in density functional calculations, an investigation of the importance of including non-linear core corrections in pseudopotential calculations, and a study of the adsorption of N_3H on aluminum surfaces.

Technical Projects Completed

Gaussian Basis Sets

A scheme for the optimization of Gaussian basis sets for use in density-functional calculations has been developed. In contrast to what is done in standard approaches, the number of primitive Gaussians used to define the basis functions is not fixed but is adjusted based on a total-energy criterion. Furthermore, all basis functions share the same set of exponents. The numerical results for the scaling of the shortest-range Gaussian exponent as a function of the nuclear charge has been explained analytically. All-electron basis sets for H, B through F, Al, Si, Mn, and Cu have been generated. They have been shown to efficiently and accurately reproduce structural properties and binding energies for a variety of clusters and molecules for both local and gradient-corrected density functionals.

Accuracy of Pseudopotentials

We have carried out a systematic investigation of the importance of nonlinear core corrections (NLCC) for accurate density-functional based pseudopotential calculations. The quality of the pseudopotential approach was assessed by making comparisons to accurate all-electron calculations. It was found that a correct description of spin-polarized states requires the use of NLCC, even for first-row atoms. The NLCC is thus essential for simulations on magnetic systems and reaction processes involving radicals. The NLCC is also essential for a realistic description of elements with more long-range core states such as alkali atoms. We have proposed a new functional form for the partial NLCC which is feasible in planewave-based calculations and we have suggested a scheme for choosing the default cutoff radius.

Adsorption and dissociation of hydrazoic acid on Al surfaces

Hydrazoic acid (N_3H) is a possible precursor for the low-temperature growth of AlN, a wide-bandgap semiconductor of industrial interest. The adsorption of N_3H on the Al(111) surface was investigated theoretically using a combination of ab initio density-functional calculations and self-consistent-charge tight-binding calculations. The adsorption and dissociation of N_3H on the Al(111) surface can be viewed as an initial step in the heteroepitaxial growth of AlN. The calculations find that the N_3H molecule dissociates with no energy barrier into N_2 and NH on the surface, which is in good agreement with experimental observations. However, while experiments provide indirect evidence for the binding of both fragments on the surface at low temperatures, the calculations find significant binding only between the NH radical and the top surface Al atoms. The source of this discrepancy remains unclear, but since N_3H-based AlN synthesis is performed at high temperatures, the possible existence of weak N_2-surface bonds is probably not crucial for understanding the growth process.

Publications

"Adsorption and dissociation of hydrazoic acid on Al(111)," D. Porezag, M. R. Pederson, and A. Y. Liu, Phys. Rev. B **61**, 13230 (2000).

"The accuracy of the pseudopotential approximation within density-functional theory," D. Porezag, M. R. Pederson, and A. Y. Liu, Phys. Status Solidi B **217**, 2190 (2000).

"Importance of nonlinear core corrections for density-functional based pseudopotential calculations," D. Porezag, M. R. Pederson, and A. Y. Liu, Phys. Rev. B **60**, 14132 (1999).

"Optimization of Gaussian basis sets for density-functional calculations," D. Porezag and M. R. Pederson, Phys. Rev. A **60**, 2840 (1999).

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. REPORT DATE *(DD-MM-YYYY)*	2. REPORT DATE	3. DATES COVERED *(From - To)*
11-08-2000	Final Technical Report	June 1997-June 2000

4. TITLE AND SUBTITLE

Final Technical Report for

"Anharmonic interactions between novel surfaces and adsorbates"

5a. CONTRACT NUMBER

5b. GRANT NUMBER

N00014-97-1-G010

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)

Liu, Amy Y.

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

Department of Physics
Georgetown University
Washington, DC 20057

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

Department of the Navy
Naval Research Laboratory
4555 Overlook Avenue
Washington, DC 20375-5326

10. SPONSOR/MONITOR'S ACRONYM(S)

NRL

11. SPONSORING/MONITORING AGENCY REPORT NUMBER

12. DISTRIBUTION AVAILABILITY STATEMENT

Approved for public release

13. SUPPLEMENTARY NOTES

14. ABSTRACT

Work was completed on three technical projects: the development of a scheme for optimizing Gaussian basis sets for use in density-functional calculations, a systematic investigation of the need to include non-linear core corrections in pseudopotential calculations, and a study of the adsorption of hydrazoic acid on aluminum surfaces.

15. SUBJECT TERMS

density-functional theory, pseudopotentials, adsorption and dissociation

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			Amy Y. Liu
U	U	U	UU	2	19b. TELEPONE NUMBER *(Include area code)* 202-687-6583